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14. ABSTRACT We describe the accomplishments of the collaborative research project performed at Ohio State University (OSU) and the University of Illinois at Urbana-Champaign (UIUC) with the support of ARO MURI grant W911NF-05-1-0414 between August 1, 2004 and July 31, 2009. Frequent interactions with Dan Rugar (IBM Almaden) have contributed significantly to our research. Our primary goal-single nuclear spin Magnetic Resonance Force Microscopy (MRFM)-calls for ultrasensitive detection as signal forces will be below 1 aN. We report several					
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## Report Title

Project W911NF-05-1-0414 Final Report: Single Nuclear Spin Magnetic Resonance Force Microscopy

### ABSTRACT

We describe the accomplishments of the collaborative research project performed at Ohio State University (OSU) and the University of Illinois at Urbana-Champaign (UIUC) with the support of ARO MURI grant W911NF-05-1-0414 between August 1, 2004 and July 31, 2009. Frequent interactions with Dan Rugar (IBM Almaden) have contributed significantly to our research. Our primary goal-single nuclear spin Magnetic Resonance Force Microscopy (MRFM)-calls for ultrasensitive detection as signal forces will be below 1 aN. We report several foundational accomplishments central to the achievement of this extraordinary sensitivity and development of methods for applying this technique to scientifically and technologically important problems. The primary milestones include but are not limited to:

1. Mitigation of sample-induced noise to enable sensitive ESR-MRFM detection of 2.3 electron spins in a 0.20 Hz bandwidth
2. Application of ultra-sensitive ESR-MRFM to the study of electron spin relaxation in spin ensembles containing of order 100 electron spins
3. Advances in development and characterization of semiconducting (Si) nanowires as next-generation ultrasensitive force detectors
4. Demonstration of high sensitivity Cu NMR measurements in technologically important layered metallic systems

We have transferred expertise and technology for high sensitivity scanned probe magnetic resonance detection to DOD and DOE labs needing state-of-the-art spin imaging capabilities; these include the Naval Research Lab (NRL) and Los Alamos National Lab. In work not directly supported by this grant, these projects advanced MRFM detected Ferromagnetic Resonance (FMR) to enable studies of submicron magnetic structures having relevance to magnetoelectronics, magnetic field sensors and the magnetic data storage.

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### List of papers submitted or published that acknowledge ARO support during this reporting period. List the papers, including journal references, in the following categories:

#### (a) Papers published in peer-reviewed journals (N/A for none)

"The Magnetic Resonance Force Microscope", P.C. Hammel and D.V. Pelekhov, Book Chapter, Handbook of Magnetism and Advanced Magnetic Materials, Helmut Kronmuller and Stuart Parkin, eds., Volume 5: Spintronics and Magnetoelectronics, John Wiley & Sons, Ltd. ISBN: 978-0-470-02217-7, (2007)

"Manipulating Spins by Cantilever Synchronized Frequency Modulation: A Variable Resolution Magnetic Resonance Force Microscope", K.C. Fong, I.H. Lee, P. Banerjee, Yu. Obukhov, D.V. Pelekhov and P.C. Hammel, Appl. Phys. Lett. 93, 012506 (2008)

"Displacement detection of silicon nanowires by polarization-enhanced fiber-optic interferometry", J.M. Nichol, E.R. Hemesath, L.J. Lauhon, R. Budakian, Appl. Phys. Lett. 93, 193110 (2008)

Number of Papers published in peer-reviewed journals: 3.00

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#### (b) Papers published in non-peer-reviewed journals or in conference proceedings (N/A for none)

Number of Papers published in non peer-reviewed journals: 0.00

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#### (c) Presentations

Presented by P. Chris Hammel:

- 1. Colloquium, Department of Physics, Penn State University, 19 February 2009
- 2. Colloquium, University of Illinois at Urbana-Champaign, IL, January 30, 2009
- 3. Presentation at the Conference on Functional Materials by Design, Los Alamos National Lab, 20 January, 2009
- 4. Invited presentation at the International Conference on Physics and Chemistry of Surfaces and Interfaces, January 9, 2009, Santa Barbara, CA
- 5. Invited presentation, International Conference on Nanoscience & Technology 21-25 July, 2008
- 6. Colloquium, Department of Physics, Northwestern University, Evanston, IL, 1 February 2008
- 7. Condensed Matter Seminar, Department of Physics and Astronomy, Michigan State University, East Lansing, MI, October 13, 2008
- 8. Invited presentation at Workshop on Frontiers of Atomic-Scale Functionality Imaging 28-30 September, 2008, Annapolis, MD
- 9. Colloquium, Department of Physics, Wayne State University, Detroit, MI, 6 December 2007
- 10. Colloquium, Department of Physics, Carnegie Mellon University, Pittsburgh, PA, 10 September, 2007
- 11. Invited Presentation at the Symposium on Nonlinear Dynamics of Nanosystems at Chemnitz, Germany, August 29, 2007.
- 12. Colloquium, Department of Physics, Kent State University, Kent, OH, 12 April, 2007
- 13. Colloquium presented at the Los Alamos National Lab Materials Colloquium, Los Alamos, NM, 21 February, 2007
- 14. Solid state seminar presented at the Department of Physics, Cornell University, Ithaca, NY, 14 November 2006.
- 15. Seminar presented at the Institute for Solid State Research of the Leibniz Institute for Solid State and Materials Research, Dresden, Germany, September 1, 2006.
- 16. Invited Presentation at the Summer School on Magnetic Resonance Force Microscopy, Ithaca, NY, June 23, 2006.
- 17. Invited Presentation at the 135th Annual Meeting of The Minerals, Metals & Materials Society (TMS), San Antonio, TX, March 13, 2006.
- 18. Colloquium, Miami University of Ohio, October 19, 2005.
- 19. Invited Presentation at EPR 2005, Columbus, OH, on 7 September 2005.

Presented by Kin Chung Fong:

- 20. Presentation at 2008 American Physical Society March Meeting March 10–14, 2008; New Orleans, Louisiana

Number of Presentations: 20.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts): 0

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts): 0

(d) Manuscripts

Number of Manuscripts: 0.00

Patents Submitted

Patents Awarded

Graduate Students

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
Kin Chung Fong	0.54
Michael Ray Herman	0.15
Inhee Lee	0.17
John Nichol	0.08
Xu Wang	0.09
Xin Zhao	0.08
<b>FTE Equivalent:</b>	<b>1.11</b>
<b>Total Number:</b>	<b>6</b>

#### Names of Post Doctorates

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
Travis Crane	0.16
Palash Banerjee	0.42
<b>FTE Equivalent:</b>	<b>0.58</b>
<b>Total Number:</b>	<b>2</b>

#### Names of Faculty Supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	National Academy Member
P. Chris Hammel	0.06	No
<b>FTE Equivalent:</b>	<b>0.06</b>	
<b>Total Number:</b>	<b>1</b>	

#### Names of Under Graduate students supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
Andrew J. Berger	0.15
Michael Roy Page	0.19
Daniel B Chait	0.08
Joshua Dylan Angelini	0.04
Eric Christopher Gingrich	0.08
<b>FTE Equivalent:</b>	<b>0.54</b>
<b>Total Number:</b>	<b>5</b>

#### Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: .....	4.00
The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:.....	3.00
The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:.....	1.00
Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):.....	0.00
Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:.....	0.00
The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense .....	0.00
The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:.....	0.00

**Names of Personnel receiving masters degrees**

<u>NAME</u>
<b>Total Number:</b>

**Names of personnel receiving PHDs**

<u>NAME</u>
Kin Chung Fong
<b>Total Number:</b>

1

**Names of other research staff**

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
Denis V. Pelekhov	0.18 No
<b>FTE Equivalent:</b>	<b>0.18</b>
<b>Total Number:</b>	<b>1</b>

**Sub Contractors (DD882)**

**Inventions (DD882)**

# Project W911NF-05-1-0414 Final Report: Single Nuclear Spin Magnetic Resonance Force Microscopy

P. Chris Hammel

*Department of Physics, The Ohio State University*

Raffi Budakian

*Department of Physics, University of Illinois at Urbana-Champaign*

(Dated: March 29, 2010)

We describe the accomplishments of the collaborative research project performed at Ohio State University (OSU) and the University of Illinois at Urbana-Champaign (UIUC) with the support of ARO MURI grant W911NF-05-1-0414 between August 1, 2005 and July 31, 2009. Frequent interactions with Dan Rugar (IBM Almaden) have contributed significantly to our research accomplishments. Our primary goal—single nuclear spin Magnetic Resonance Force Microscopy (MRFM)—calls for ultrasensitive detection as signal forces will be below 1 aN ( $10^{-18}$  N). We report several foundational accomplishments central to the achievement of this extraordinary sensitivity and development of techniques for applying this technique to scientifically and technologically important problems:

- Mitigation of sample-induced noise to enable sensitive ESR-MRFM detection of 2.3 electron spins in a 0.20 Hz bandwidth
- Application of ultra-sensitive ESR-MRFM to the study of electron spin relaxation in spin ensembles containing of order 100 electron spins
- Advances in development and characterization of semiconducting (Si) nanowires as next-generation ultrasensitive force detectors
- Reduction of intrinsic dissipation in Si nanowires through surface passivation; this establishes the foundations for low noise force detection
- Measurement of sample surface induced noise in ultra-sensitive Si nanowire force detectors; these results reveal superior performance relative to conventional ultra-soft cantilevers
- Demonstration of high sensitivity Cu NMR measurements in technologically important layered metallic systems
- Work on ultrasensitive displacement detection for sensitive force detectors including SQUID based displacement readout

We have transferred expertise and technology for high sensitivity scanned probe magnetic resonance detection to DOD and DOE labs needing state-of-the-art spin imaging capabilities; these include the Naval Research Lab (NRL) and Los Alamos National Lab. In work not directly supported by this grant, these projects advanced MRFM detected Ferromagnetic Resonance (FMR) to enable studies of submicron magnetic structures having relevance to magnetoelectronics, magnetic field sensors and the magnetic data storage.

We also present our results of MRFM sample characterization measurements of materials obtained using a Bruker Electron Spin Resonance (ESR) Spectrometer purchased with the support of ARO DURIP grant W911NF-07-1-0305. These results have already been presented in the final report for the ARO DURIP grant. We report them here as well because the ARO DURIP grant provided funds for instrumentation only and the sample characterization was performed by personnel supported by the this ARO MURI grant W911NF-05-1-0414.

## INTRODUCTION

We report results from our collaborative research program in which techniques for ultrahigh sensitivity MRFM nuclear spin detection were developed and MRFM was applied to scientific and technological problems of importance to DoD, industry and society. Our goal of single nuclear spin detection calls for unprecedented sensitivity in the mechanical detection of the weak signals generated by individual spins, nuclear or electronic. This problem is particularly acute when it must be met in the context of samples and materials of interest because they introduce a range of additional challenges. Primary amongst

these are undesired interactions between the sample and the sensitive force detector. The primary requirement is for excellent sensitivity in the particular experimental context. We report advances central to achieving this extraordinary sensitivity, and to achieving high sensitivity in the application of this powerful technique to problems of scientific and technological interest achieved during the time period covered by this report.

- Mitigation of sample-induced noise enabled sensitive ESR-MRFM detection of 2.3 electron spins in a 0.20 Hz bandwidth

- Application of the ultra-sensitive ESR-MRFM to the study of electron spin relaxation in spin ensembles containing of order 100 electron spins
- Important advances in development and characterization of semiconducting (Si) nanowires as next-generation ultrasensitive force detectors
- Reduction of intrinsic dissipation in Si nanowires through surface passivation; thus providing foundations for low force noise detection
- Measurement of sample surface induced noise in ultra-sensitive Si nanowire force detectors; these results revealed superior performance relative to conventional ultra-soft cantilevers
- Demonstration of high sensitivity Cu NMR measurements in technologically important layered metallic systems
- Work on ultrasensitive displacement detection for sensitive force detectors including SQUID based displacement readout

We also present MRFM sample characterization measurements obtained using a Bruker Electron Spin Resonance (ESR) Spectrometer purchased with the support of ARO DURIP grant W911NF-07-1-0305. These results have already been presented in the final report for the ARO DURIP grant. We report them here as well because the ARO DURIP grant provided funds for instrumentation only and the sample characterization was performed by personnel supported by the this ARO MURI grant W911NF-05-1-0414.

### ULTRASENSITIVE ESR/MRFM IN $\text{SiO}_2$

We have demonstrated 2 electron spin sensitivity in a sample of  $\gamma$ -irradiated  $\text{SiO}_2$ . This is an excellent sample for developing and diagnosing ultrahigh sensitivity MRFM. To improve reproducibility and allow benchmarking we have used a sample obtained from Dan Rugar (IBM-Almaden) that allows us to compare results obtained by his group, ours and the UIUC group. The electronic spins are associated with  $\text{E}'$  centers that result from  $\gamma$ -irradiation of a polished silica crystal. Here we describe high sensitivity experiments performed in a sample having spin density  $\sim 7 \times 10^{17} \text{ cm}^{-3}$ .

A schematic diagram of the experimental apparatus is shown in Fig. 1. It consists of an ultrasoft cantilever, fabricated in collaboration with IBM and UIUC, with an attached SmCo micromagnetic probe tip (shaped by Focused Ion Beam [FIB] machining to  $\sim 1 \mu\text{m}$  characteristic dimensions) mounted on its tip. This tip generates a field gradient exceeding 2 G/nm. A microwave magnetic field is generated by a superconducting microcoil that we developed in collaboration with IBM and UIUC. A homogeneous external magnetic field  $B_{\text{ext}}$  is generated

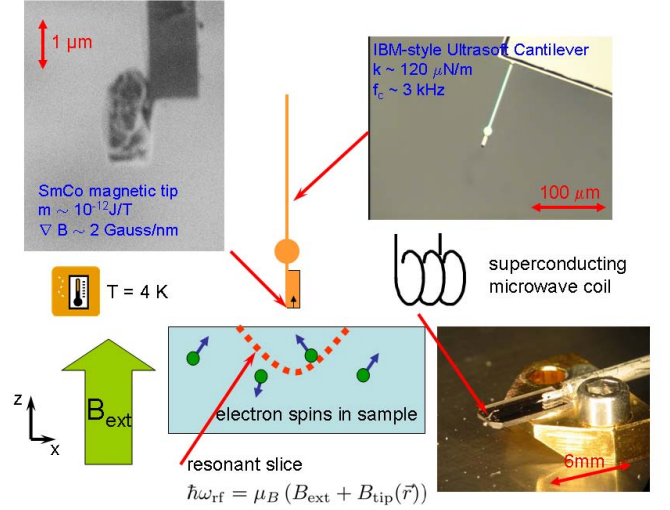


FIG. 1: The experimental setup used in the ultrahigh sensitivity ESR MRFM measurements. An ultrasoft cantilever, developed as a collaboration between IBM, UIUC and OSU, is used. The SmCo probe magnet with characteristic dimensions of  $1 \mu\text{m}$  has been fabricated by FIB machining. The typical achievable field gradient is 2 G/nm.  $rf$  field is generated by a superconducting microcoil developed as a collaboration between IBM, UIUC and OSU. We use the magnitude of the applied external magnetic field  $B_{\text{ext}}$  to control the spatial location of the resonant slice relative to the probe magnet. The distance between the probe magnet and the sample surface is  $D$ .

by a superconducting solenoid. This field controls the spatial location of the resonant slice relative to the probe magnet. The overlap of the resonant slice with the sample which determines the number of spins participating in the experiment is adjusted by highly precise control of the probe-sample separation  $D$  (Fig. 1).

The MRFM signal is encoded using the interrupted OSCAR spin manipulation technique [1]. The signal was recorded as a function of frequency offset  $f$  from the interrupt frequency  $\omega_m$ . The spectral density of the statistical polarization signal  $S_f(f)$  exhibits the expected Lorentzian dependence on frequency:

$$S_f(f) = N \frac{4\tau_m \delta f^2}{1 + (2\pi f)^2 \tau_m^2},$$

where  $\tau_m$  is the signal lifetime. Using this expression and knowledge of the probe field gradient we can estimate the total number of spins generating the signal.

### Wide bandwidth, highly sensitive detection of statistically polarized spins in $\text{SiO}_2$

Fig. 2 shows data obtained from signals from a systematically reduced number of spins obtained by increasing the  $D$  increases 450 nm to 1050 nm, and demonstrating a

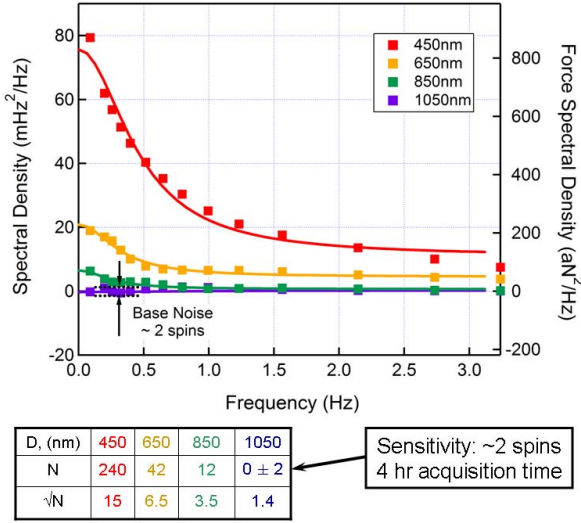


FIG. 2: Signal from statistically polarized  $E'$  centers in  $\text{SiO}_2$  recorded at several probe-sample distances  $D$ . The spectral density of the statistical polarization signal  $S_f$  is shown as a function of frequency offset from the OSCAR interrupt frequency  $\omega_m$ . The total number of spins generating the signal is known from the frequency dependence of  $S_f$  and knowledge of the probe field gradient. As the slice is retracted from the sample by increasing  $D$  from 450 nm to 1050 nm, the number of spins contained in the slice decreases from 240 to  $\sim 2$  while the noise remains constant. The blue curve shows a **two electron spin** sensitivity MRFM signal obtained after four hours of signal averaging.

minimum detectable signal decreasing from  $240 \mu_B$  ( $\mu_B$  is the Bohr magneton) to  $\sim 2 \mu_B$ . The blue curve shows a two electron spin sensitivity MRFM experiment. However, in this experiment, this data required *four hours of signal averaging*. Such slow data acquisition is unacceptable if a significant amount of information is to be acquired.

By means of sensitivity improvements described below, our current microscope is able to detect fluctuations of statistically polarized electron spins [1, 2] with 2.3 electron spin sensitivity in a data acquisition bandwidth of 0.2 Hz (nearly *real time* data acquisition compared to the four hours of signal averaging that was achieved previously). Figure 3 shows signals that we obtain from a sample of  $\text{SiO}_2$  that has been  $\gamma$ -irradiated to generate unpaired electron spins (*i.e.*,  $E'$  centers) having long spin lifetimes (exceeding 1 second). The ESR signal is due to the force that statistically polarized spins exert on the cantilever [1], and is evident through the shift in the resonant frequency of the cantilever it induces. This signal is generated by periodically rotating the statistical spin polarization such that it is either in-phase or  $180^\circ$  out-of-phase with the lock-in reference. The signal evident in the in-phase channel in Fig. 3 arises from 13.1 net spins. By controlling signal phase, we ensure that only detector

noise contributes to the quadrature channel. The standard deviation of the cantilever frequency shift in the quadrature channel is 2.0 mHz which corresponds to the measurement noise floor of **2.3 net electron spins**. By increasing averaging times to approximately 9 hours, the signal from an individual spin can be detected with unity signal to noise ratio.

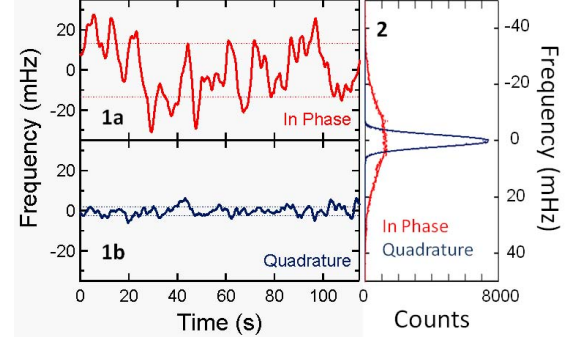


FIG. 3: Statistical polarization ESR signal  $E'$  centers in a  $\gamma$ -irradiated silica sample recorded by means of our ultra-sensitive MRFM. (1) Observation of real-time spin fluctuations. Lock-in traces of statistical polarization signals in the in-phase (Fig. 1a) and quadrature (Fig. 1b) channels are taken with 0.20 Hz bandwidth (0.87 s lock-in time constant); the spin correlation time is about 3.0 s. (2) Spin fluctuation histogram from the time traces in Panel 1. The solid lines are best fits of Gaussian functions to the data. The standard deviations of the in-phase and quadrature channels are 13 and 2.0 mHz respectively; the latter characterizes the detector noise corresponds to **2.3 net electron spins**. The spin signal, which contributes to the in-phase signal alone, corresponds to  $\sim 13.1$  net spins.

## ESR-MRFM EXPERIMENTS ON FEW ELECTRON ENSEMBLES

We have applied our ultra-sensitive MRFM-detected electron spin detection to nanoscopic measurement of local environments as reflected in spin dynamics of small,  $\sim 100$  spin ensembles.

### Spin relaxation in small ensembles

Phosphorus doped silicon (Si:P) has tremendous technological importance as the workhorse of consumer electronics. The ability to microscopically image and study the properties of individual donors would substantially impact this industry. Furthermore this material has substantial interest arising from the potential for using the spin of the electron in classical and quantum information processing. For this application tools for studying and manipulating the individual spins buried in silicon will

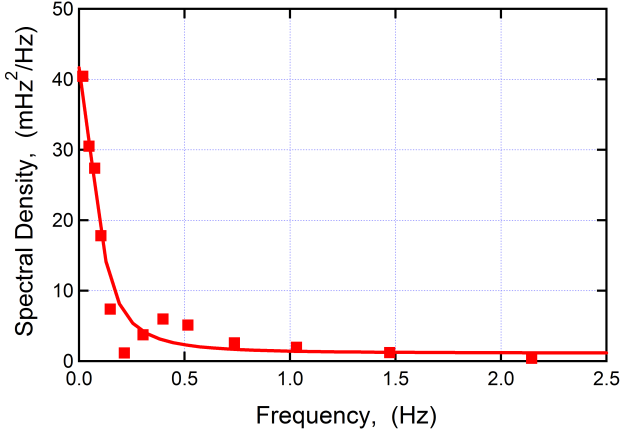


FIG. 4: Signal arising from statistically polarized electronic moments in Si:P obtained at a probe-sample separation  $D \approx 600$  nm; in this case the estimated probe field gradient is  $\sim 0.1$  G/nm. The estimated signal life time  $\tau_m$  is  $1.3 \pm 0.5$  s. The signal shown originates from an ensemble  $\sim 80$  electron spins, this corresponds to net number of polarized  $\sim 9$  net spins.

be essential, a role that MRFM could be uniquely well suited to fill.

The sample used for our ESR/MRFM Si:P experiments is a custom grown SOI wafer consisting of a thick Si substrate with  $9 \mu\text{m}$  thick Si top layer doped with P to concentration of  $\sim 1.0 \times 10^{16} \text{ cm}^{-3}$ . The P doped layer is separated from the substrate by a  $2 \mu\text{m}$  layer of  $\text{SiO}_2$ .

The hyperfine interaction between the electronic spins and the both  $^{31}\text{P}$  and  $^{29}\text{Si}$  nuclear moments presents both a challenge and opportunities for spin electronic applications of Si:P. NMR experiments on these nuclear spins will provide essential insights for these technologies and offer excellent scientific opportunities as well. Electron Nuclear Double Resonance (ENDOR) will enable direct study of both individual nuclear spins and their interaction with the electronic spins.

In addition to their intrinsic importance, these experiments will underlie the application of ESR/MRFM to semiconducting materials and particularly for study of donors in semiconductors. Successful experimentation in these systems will be built upon thorough understanding of sample induced noise phenomena and means for their mitigation as described in the following section and summarized in Fig. 7.

In our first experiments we used a sample prepared out of isotopically enriched  $^{28}\text{Si}$  to remove the effect of the  $^{29}\text{Si}$  nuclear spins on the electron spin lifetime and better understand the intrinsic electronic spin properties. Results are presented in Fig. 4 which shows the spectral density of the statistical polarization signal  $S_f$  as a function of the frequency offset from the interrupt frequency  $\omega_m$  in the interrupted OSCAR spin manipulation protocol. An analysis similar to that used for the MRFM

signals from E' centers in  $\text{SiO}_2$  shows that this signal originates from an ensemble containing  $\sim 80$  electron spins so the average spin polarization is 9. For this experiment the probe-sample separation was  $D \approx 600$  nm; at this distance the estimated strength of the magnetic field gradient of the probe was only  $\sim 0.1$  G/nm.

### Spin relaxation in small ensembles

Electron spins are relaxed by local magnetic field fluctuations so relaxation rate measurements provide microscopic information regarding dynamics of the spin's crystalline, molecular and magnetic environment, and hence is a powerful probe of material properties. Conventional ESR requires relative large ensembles of electron spins (typically  $\gtrsim 10^9$ ) and hence provides signals that are superpositions over large volumes. Our capability for ultra-sensitive ESR detection outlined in Sec. enables us to measure electron spin relaxation in ensembles of less than 100 electron spins.

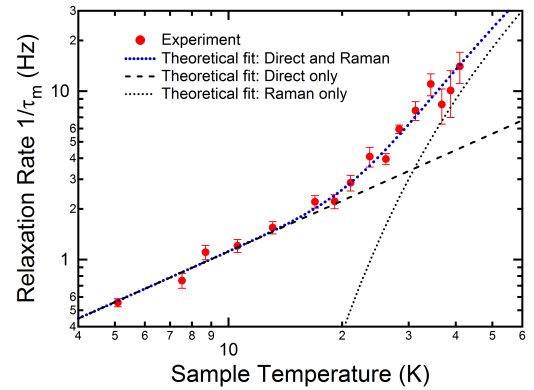


FIG. 5: Electron spin relaxation rate  $1/\tau_m$  of E' centers in  $\gamma$ -irradiated silica sample shown as a function of sample temperature  $T$ . The fit to experimental data using Eq. 1 shows that at temperatures below  $T \approx 16$  K electron spin relaxation occurs primarily due to the direct phonon absorption/emission, while a two-phonon Raman process dominates spin relaxation at higher temperatures. The contributions of the direct and Raman relaxation processes are shown.

In Fig. 5 we show our measurements of the temperature dependence of the electron spin relaxation rate  $1/\tau_m$  of E' centers in  $\gamma$ -irradiated  $\text{SiO}_2$ . Our experimental data are well described by conventional electron spin relaxation theory [3–5]. Two distinct phonon-based mechanisms are responsible for the two regimes of temperature dependent spin relaxation evident in Fig. 5; these data are described by the following temperature dependence:

$$\frac{1}{\tau_m} = AT + C \frac{e^{T_{\text{mode}}/T}}{[e^{T_{\text{mode}}/T} - 1]^2}, \quad (1)$$

Here  $A$  is a fit parameter describing spin relaxation due to direct absorption/emission of a phonon whose frequency matches the applied  $rf$  frequency  $\omega_{rf}$ , and  $C$  describes the scale of spin relaxation by a two-phonon Raman process.  $T_{\text{mode}}$  gives the energy scale for vibrational quanta of an  $E'$  structural defect expressed in units of temperature. Fig. 5 shows that the direct relaxation mechanism dominates at temperatures below  $T \approx 16$  K while the Raman process dominates at higher temperatures. The parameter  $T_{\text{mode}} = 124 \pm 18$  K measures the mechanical resonant frequency of the  $E'$  structural defect  $f_{E'} = (k_B T_{\text{mode}})/h \approx 2.6 \times 10^{12}$  Hz in good agreement with published data [3–5].

These measurements demonstrate the emergent capability for nanoscopic imaging of electron spin dynamics in addition to static spin properties such as density. Further, the results shown in Fig. 5 confirm that the electron spin relaxation is due to intrinsic relaxation mechanisms; importantly *our MRFM detection protocol does not degrade spin lifetime*.

The ability to control spin relaxation time by varying sample temperature provides an important tool for optimizing Signal-to-Noise Ratio (SNR) in MRFM. Degen *et al.*, have shown [6] that for a given noise spectral density  $S_{\text{noise}}$  and spin signal strength  $\sigma_{\text{spin}}$ , the SNR of the statistical polarization MRFM measurement is optimal for a particular value of the spin memory randomization time:  $\tau_r = S_{\text{noise}}/(\sqrt{2}\sigma_{\text{spin}}^2)$ . This is because each independent measurement improves the SNR and  $\tau_r$  defines the number of independent measurements that can be acquired during the total measurement time  $\tau_{\text{tot}}$ . In the case where the spin relaxation time  $\tau_m$  is long and cannot be controlled by other means,  $\tau_r$  can be reduced by artificially randomizing spins using  $rf$  pulses [6].  $\tau_m$  sets an upper limit on  $\tau_r$ , and modifying  $\tau_m$  directly through temperature provides a key alternative mechanism for optimizing experimental SNR.

## SI NANOWIRE CANTILEVERS

Single nuclear spin MRFM detection will require further advances in sensitive force detection, so we are investigating silicon nanowires (SiNW) as potential sub-attonewton force detectors. Nanowires hold great promise for ultra-high sensitivity force detection because they can be synthesized epitaxially with nanometer scale lateral dimensions and lengths of tens of microns. Their high aspect ratio, low mass, and corresponding low mechanical stiffness and high flexural frequency are ideally suited for sensitive force detection. The force sensitivity of a SiNW could be quite high. A nanowire having a 20 nm diameter and 15  $\mu\text{m}$  length should theoretically have a stiffness of 2  $\mu\text{N}/\text{m}$  and 120 kHz fundamental flexural frequency. At a temperature of 4 K, the force sensitivity of such a nanowire would be 0.3 aN/ $\sqrt{\text{Hz}}$  assuming

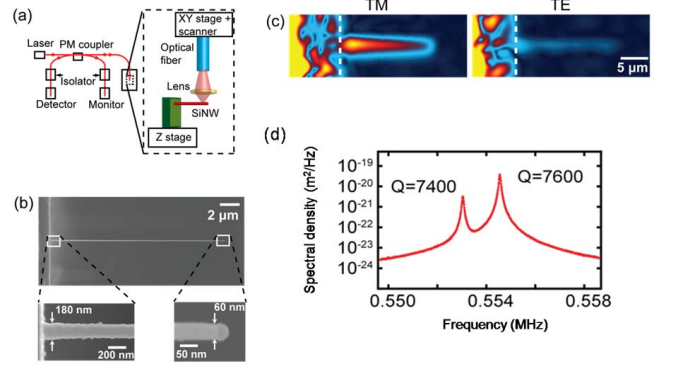


FIG. 6: (a) Schematic of the scanning interferometer, (b) SEM image of an individual single-crystal SiNW, (c) optical image of the SiNW shown in (b) obtained using the scanning fiber optic interferometer. Images were taken with transverse magnetic (TM) and (TE) polarizations. For (TM) polarized light, the scattered intensity increases by nearly 50 fold. (d) Thermally excited frequency spectrum of the SiNW shown in (b). The two peaks correspond to the two lowest flexural modes of the SiNW.

a modest quality factor of 5000. Two of the key challenges to using SiNW for force detection are (1) synthesis of nanowires having low dissipation, i.e., high mechanical  $Q$ , and (2) the ability to detect displacement of nanowires with sub- $\text{\AA}$  displacement sensitivity. Through a collaboration with Professor Lincoln Lauhon (Department of Materials Science and Engineering, Northwestern University), an expert in fabricating semiconductor nanowires, we are exploring using single-crystal SiNW as cantilevers for MRFM. An array of vertical nanowires that are 20–30 nm in diameter and 10–20  $\mu\text{m}$  long will be grown epitaxially from silicon posts using the vapor-liquid-solid (VLS) method. Controlled registry to the substrate will be achieved by patterning the metal catalyst pads, using electron-beam lithography. Fig. 6b shows an SEM image of a single-crystal SiNW used in the reported experiments.

## Polarization-Enhanced Fiber Optic Interferometry

Optical detection is the most widely used and versatile technique for displacement detection of cantilevers in scanning probe applications. Optical interferometry, in particular, offers excellent displacement sensitivity in the  $10^{-13}$ – $10^{-14}$  m/ $\sqrt{\text{Hz}}$  range with low incident optical power 0.1–1  $\mu\text{W}$ . Until recently, optical detection was primarily used with cantilevers having dimensions in the micron range. It is generally believed that as the size of the cantilever becomes smaller than the wavelength of light, the scattered intensity from the cantilever decreases significantly and sacrifices displacement sensitivity. It is possible, however to exploit the polariza-

Base diameter (nm)	Tip diameter (nm)	Length ( $\mu\text{m}$ )	$k$ ( $\mu\text{N/m}$ )	$\omega_0/2\pi$ (kHz)	$Q_{\text{native}}$ ( $10^3$ )	$Q_{\text{passiv}}$ ( $10^3$ )	$\Gamma_{\text{native}}$ ( $10^{-15}$ kg/s)	$\Gamma_{\text{passiv}}$ ( $10^{-15}$ kg/s)	$S_f^{1/2}$ ( $10^{-13}$ N/ $\sqrt{\text{Hz}}$ )
175	60	17.7	677	533(555)	7.5(7.5)	9.5(9.0)	26(26)	21(22)	18(19)
148	50	17.4	380	497(498)	7.0(7.0)	9.0(8.0)	17(17)	15(15)	15(16)
153	48	18.0	390	347(347)	7.0(6.5)	9.0(9.0)	26(28)	20(26)	18(18)
46	46	12.9	66	265(281)	4.0(4.5)	7.0(7.5)	10(8)	6(5)	10(9)
44	44	14.4	28	208(213)	4.0(4.0)	10.0(8.5)	5(5)	2(3)	6(7)

TABLE I: Mechanical properties of the fundamental modes for selected SiNWs. Parameters for the higher frequency mode are given in parentheses. The subscripts "native" and "passiv" denote before and after passivation. For force sensitivity calculations,  $\Gamma_{\text{passiv}}$  values were used.

tion of light to significantly increase the scattered intensity and maintain high displacement sensitivity. We have constructed a polarized scanning fiber optic interferometer that focuses light to a  $2\ \mu\text{m}$  spot size and can locate individual self-standing silicon nanowires grown on a Si(111) surface and interferometrically detect the displacement of the nanowire. The instrument, shown schematically in Fig. 6a, uses  $\lambda = 1060\ \text{nm}$  light. Light that is polarized along the axis of the nanowire should have nearly a factor of 100 larger reflection coefficient than light that is polarized perpendicular to the axis of the nanowire. The epitaxial single-crystal SiNWs are grown in the group of Professor Lauhon at Northwestern University using the vapor-liquid-solid process. The SiNWs are 50–60 nm in diameter and 10–15  $\mu\text{m}$  in length. Fig. 6 shows a schematic of the instrument along with data obtained from individual SiNWs. Using this instrument, we have achieved high displacement sensitivity  $\sim 5 \times 10^{-13}\ \text{m}/\sqrt{\text{Hz}}$  range with 15  $\mu\text{W}$  of 1550 nm light incident on the nanowire [7].

### Improving Mechanical Properties of SiNWs by Surface Passivation

One of the primary purposes of the polarization enhanced optical interferometer described above is to characterize the mechanical dissipation of SiNWs since this will ultimately define the sensitivity of the instrument, and to seek ways to decrease it. The intrinsic mechanical dissipation is given by  $\Gamma_c = k/\omega Q$ , where  $k$ ,  $\omega$ , and  $Q$  are the cantilever spring constant, frequency and quality factor respectively. We have attempted to reduce dissipation in SiNWs by surface passivation. Table I shows tabulated data for cylindrical and tapered SiNWs. Each nanowire was measured at room temperature first without any surface treatment, and then after placing the SiNW into a vapor HF chamber to remove the native oxide and hydrogen passivate the silicon surface. For both kinds of SiNWs studied, we find that the quality factors improved by about a factor of 2.0 after hydrogen passi-

vation. The lowest mechanical dissipation observed after passivation was  $2 \times 10^{-15}\ \text{kg/s}$ . It is important to point out that this dissipation value is nearly  $50\times$  smaller than the dissipation of the traditional ultrasensitive cantilevers (similar to shown in Fig. 1) used for single-electron spin detection [2]. This will significantly improve detection sensitivity. The low mass and high frequency of SiNW cantilevers also make them well-suited for Larmor spin detection, where the spin precession would directly drive the resonant cantilever at the Larmor frequency.

### NON-CONTACT FRICTION: MITIGATION THROUGH SURFACE PREPARATION

Single electron spin electron spin resonance has been demonstrated using MRFM; this opens a wide range of newly feasible applications to scientific and technological applications. We are pushing toward single nuclear spin MRFM detection and developing the techniques, hardware and knowledge base to apply this capability to important scientific and technological challenges. A fundamental technological hurdle for single nuclear spin MRFM is control and mitigation of the detection noise induced by the sample itself.

We have studied non-contact friction using ultra-soft Si cantilevers; recently we have extended these experiments to include the high performance SiNW cantilevers. We present both of these results here.

#### Non-contact friction studies in ultra-soft cantilevers

Maximizing the sensitivity of the MRFM in the actual context of the needed measurement is at the heart of these advanced applications. One measure of sensitivity is signal-to-noise ratio (SNR). The signal strength is determined by the dipolar coupling between the micromagnetic tip on the MRFM cantilever, and the spin magnetic moment of the sample; in our magnetic field gradients exceeding  $2 \times 10^5\ \text{T/m}$ , a single electron spin exerts a 2

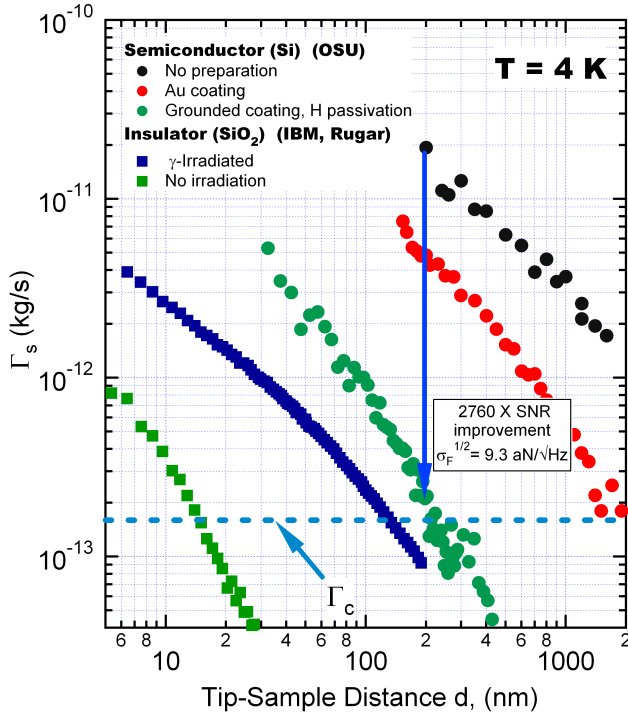


FIG. 7: Sample induced dissipation due to non-contact friction between an ultrasensitive cantilever and a surface as a function of the separation between the probe tip and the sample surface. The surface noise is larger for semiconducting samples (Si) than for insulating ( $\text{SiO}_2$ ) samples. Passivation of the silicon surface in a buffered oxide etch (hydrofluoric acid) followed by coating with 20 nm gold layer and grounding of the coating film reduces surface dissipation  $\Gamma_s$  from  $1.93 \times 10^{-11}$  kg/s to  $2.11 \times 10^{-13}$  kg/s at a probe-sample separation of at 200 nm. We believe that hydrogen passivation removes trapped charges from the Si surface thus reducing dissipation and improving the signal-to-noise (SNR) ratio

aN force. In addition to thermomechanical noise, surface noise becomes important once the tip closely approaches the sample surface. Using specially designed **ultrasoft cantilevers** (fabricated in collaboration with D. Rugar, IBM and R. Budakian, UIUC) we have reached cantilever noise level as low as 6–10 aN/ $\sqrt{\text{Hz}}$ .

Upon close approach, the sample noise exceeds thermal noise thus reducing detection sensitivity. Sample induced dissipation is not well understood, but it is generally attributed to probe interactions with charge either on the surface or within the sample. This mechanism is especially significant for materials with surfaces that might have large number of charged defects such as P doped Si. We have investigated the dependence of the surface dissipation on the probe-sample distance and methods of Si surface preparation. The total dissipation is the sum of sample dissipation  $\Gamma_s$  and intrinsic cantilever dissipation  $\Gamma_c$  giving a total force noise power density  $\sigma_{\text{noise}} = 4k_B T(\Gamma_s + \Gamma_c)\delta\nu$ , where  $k_B T$  is the thermal en-

ergy. The measured sample dissipation  $\Gamma_s$  is shown in Figure 7. Hydrogen passivating and coating the sample reduced the dissipation  $\Gamma_s$  at 200 nm separation from  $1.93 \times 10^{-11}$  kg/s to  $2.11 \times 10^{-13}$  kg/s. We believe hydrogen passivation reduces the trapped charge density on the Si surface thus reducing dissipation and improving  $\text{SNR} = (\sigma_{\text{sig}}/\sigma_{\text{noise}})^2$ , where  $\sigma_{\text{sig}}$  is the signal power density. The SNR of a cantilever with intrinsic dissipation  $\Gamma_c = 1.6 \times 10^{-13}$  kg/s improves by over  $\sim 2700$  times. This is a very significant improvement that reduces the data acquisition time by  $\sim 2700$  times thus making ultrahigh sensitivity experiments possible.

### Non-contact friction measurements in Si nanowire cantilevers

In order to achieve the high field gradients required for ultrasensitive spin detection, the nanomagnet must be brought close to the sample. For single nuclear spin detection, where field gradients in excess of  $10^7$  T/m are required, the spacing should be of order 10 nm. In non contact friction measurements made with low frequency ultrasoft cantilevers, it has been observed that when the tip to surface spacing becomes smaller than about 100 nm, surface induced dissipation begins to dominate the intrinsic dissipation of the cantilever. The force noise  $S_F = 4k_B T\Gamma$  experienced by the cantilever is determined by the total cantilever dissipation  $\Gamma = \Gamma_c + \Gamma_s$ , where  $\Gamma_s$  is the surface-induced dissipation. Fig. 8 compares surface-induced dissipation in ultrasoft, low frequency cantilevers with micron-size tips to that seen in a high frequency Si nanowire cantilever (SiNW) with a 50 nm tip diameter.

At 300 K, the surface induced force noise for the SiNW cantilever is approximately  $15\times$  lower than the low frequency cantilever. The lower force noise observed for SiNW cantilever could be due to the smaller tip size as well as the higher operating frequency. The underlying surface interactions that give rise to mechanical dissipation are still not well understood. Overall, the total dissipation  $\Gamma$  observed for the 1 MHz SiNW cantilever 10 nm away from the surface is approximately  $30\times$  lower at 300 K than the lithographically fabricated low frequency cantilevers at 4 K. This large gain in sensitivity opens up the possibility of sub-attoneutron MRFM measurements close to a surface. Currently, we are constructing a low temperature SiNW-MRFM microscope which will operate at 4K for high sensitivity nuclear spin measurements.

### SQUID BASED DISPLACEMENT READOUT

We have made significant progress in fabricating of a SQUID magnetometer for MRFM cantilever displacement readout. Fig. 9 shows a schematic of our design along with images of the devices that we have fabricated.

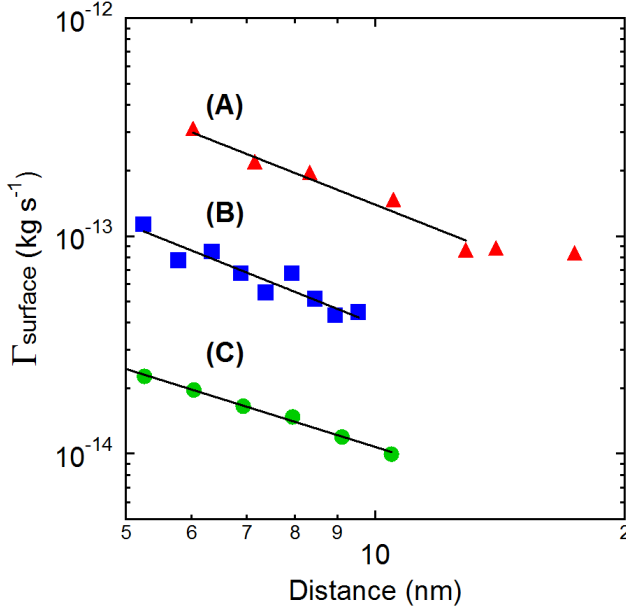


FIG. 8: (A) and (B) Surface-induced dissipation near the surface of Au(111) obtained by Stipe *et al.*[8] using a low frequency ultrasoft cantilever having a frequency  $f=3.86$  kHz, spring constant  $k = 330 \mu\text{N/m}$  and a quality factor of  $Q=17,200$  at 4 K. Data shown in (A) and (B) were taken at 300 K and 4 K, respectively. (C) surface-induced dissipation obtained near the surface of Si(100) using a SiNW cantilever having a frequency  $f = 1.08$  MHz,  $k = 600 \mu\text{N/m}$ , and  $Q = 5,400$  at 300 K.

The mechanical portion of the device consists of a 100-nm thick doubly clamped suspended silicon nitride beam that is  $22 \mu\text{m}$  long. The beam is covered by a 40-nm thick layer of Nb, which is part of a larger Nb washer. During operation, a magnetic field in the 10–30 mT range will be applied in the plane of the washer and perpendicular to the axis of the cantilever. As the cantilever is displaced out of the plane of the substrate by a distance  $\Delta x$ , a magnetic flux given by  $\Delta\phi \approx l\Delta x B/2$  will enter the Nb washer, where  $l$  is the length of the cantilever and  $B$  is the external magnetic field. The flux will then be coupled to an off-chip dc SQUID through a flux transformer. The output of the SQUID will be amplified using an ultra-low-noise SQUID array amplifier before going to the room electronics. The displacement sensitivity of our design is given by  $S_x = 2S_\phi/lB$ , where  $S_x$  and  $S_\phi$  are the displacement and flux noise spectral density, respectively. For  $S_\phi = 3 \times 10^{-6} \phi_0/\sqrt{\text{Hz}}$  of our SQUID at 4 K, our expected displacement sensitivity is  $S_x = 3 \times 10^{-14} \text{ m}/\sqrt{\text{Hz}}$ , representing more than a factor of 10 improvement in displacement sensitivity over optical detection. The current doubly clamped cantilever design is meant to be a proof of concept for SQUID-based displacement detection. After testing this new detection

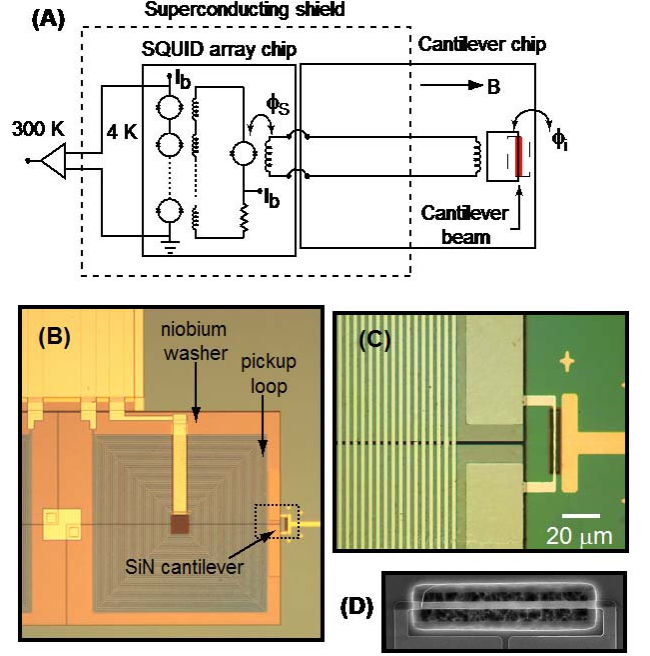


FIG. 9: (A) Schematic diagram of the SQUID detected cantilever design. (B) and (C) Optical image of the superconducting Nb pickup loop and SiN cantilever fabricated by the UIUC group. The doubly clamped beam is expected to have a resonant frequency of 2 MHz and a stiffness of 3 N/m. (D) SEM image of the suspended beam.

scheme, we will integrate this detection technology with the ultra-sensitive cantilevers to be used for MRFM.

## HIGH SENSITIVITY NMR

The application of ultrahigh sensitivity NMRFM to general samples presents new challenges that must be overcome to ensure that the advances in MRFM sensitivity can contribute to scientific and technological advances. Building on our earlier NMR detection of  $^{19}\text{F}$  spins in  $\text{CaF}_2$  we have conducted  $^{65}\text{Cu}$ ,  $^{63}\text{Cu}$  NMR studies for studies of interface phenomena in multilayered magnetic systems. Our experiments were conducted at low temperatures (4–30 K) using a micron-scale NdFeB spherical probe magnet mounted on a commercial  $\text{Si}_3\text{N}_4$  cantilever with spring constant  $k \sim 0.01 \text{ N/m}$ .

The nuclear spins were manipulated using fast adiabatic inversion. The frequency of the applied *rf* magnetic field is modulated at the resonant frequency of the cantilever to resonantly drive it. For sufficiently strong *rf* magnetic field (meeting the adiabatic condition) the nuclear spins are coherently rotated. If the *rf* magnetic field is too weak coherent manipulation of the polarized spin ensemble is impossible. We used a tunable *rf* tank circuit with a microcoil inductor for generating the intense os-

cillating  $rf$  field  $H_1$ . The  $H_1$  intensity was measured by measuring the nutation of  $^{19}\text{F}$  spins (Fig. 10b) by the  $rf$  field. The oscillations of the MRFM signal intensity were measured as a function of the duration of the applied  $rf$  pulse. The period ( $= 2\pi\gamma H_1$  where  $\gamma$  is the nuclear gyromagnetic ratio) of these oscillations gives  $H_1 = 7.8$  G obtained with 100 mW of applied  $rf$  power. We have also measured the nuclear spin relaxation time  $T_1$  (Fig. 10a); the result,  $T_1 \approx 17$  s, agrees well with conventional NMR measurements on the same sample.

Our experiments on  $^{65}\text{Cu}$ ,  $^{63}\text{Cu}$  spins in a thin Cu foil sample (Fig. 11a) built on these results. Because  $^{63}\gamma < ^{19}\gamma$  a proportionally larger  $H_1$  is needed to meet the adiabatic condition; this was achieved with a novel tuned circuit with two microcoils that allowed the sample coil to be small (volume  $V$ ) hence increasing  $H_1 \sim \sqrt{P/V}$  for a given power. The larger field was confirmed in the  $^{63}\text{Cu}$  nutation experiment that demonstrated  $H_1 = 25$  G (Fig. 11b). The spin sensitivity was  $10^5$  spins. We are now engaged in NRMFM experiments on Co-Cu bilayers; a microscope able to study the buried Cu/Co interface would be a powerful tool for understanding and developing magnetoelectronics and the ability to perform high sensitivity experiments in conductive samples will open many potential systems to MRFM study. Even with current sensitivity we can detect nuclear spins in sub-nm layer adjacent to the interface with lateral resolution of a few micrometers.

One of the key sources of non-thermal noise is the spurious coupling of the MRFM cantilever to the applied  $rf$  radiation. To understand the roles of heating mechanisms for this coupling we have explored the dependence of spurious coupling on the thermal conductivity of the cantilever. In particular we have compared low thermal conductivity  $\text{Si}_3\text{N}_4$  to higher thermal conductivity Si and found  $\text{Si}_3\text{N}_4$  to be substantially worse indicating that heating is an important contributor to this effect. We are considering moving to the ultrasoft Si cantilevers of the same type as the one used in our ultrasensitive ESR experiments. Another mechanism of spurious cantilever excitation is electrostatic coupling between the cantilever and the  $rf$  coil. We are developing approaches to shielding the coil  $rf$  source to reduce its effect on the cantilever.

## ELEMENTS OF ULTRAHIGH SENSITIVITY MRFM

The ultrahigh sensitivity MRFM results enabling **two electron spin** detection discussed earlier were built upon several technical advances implemented over the course of the last year. Many of these advances have grown out of close collaboration between the OSU, UIUC and IBM-Almaden groups; this collaboration is summarized below.

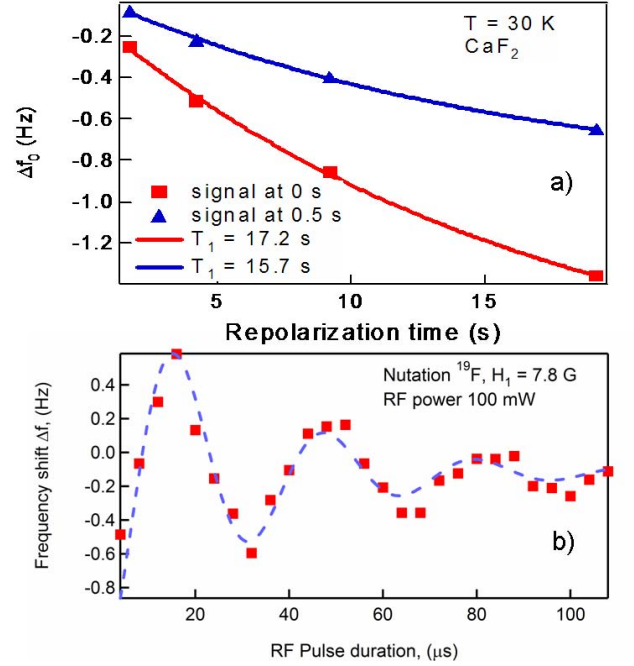


FIG. 10: Characterization and optimization of the NRMFM instrument using  $^{19}\text{F}$  NMR in  $\text{CaF}_2$ : a)  $T_1$  measurement at  $T = 30$  K. Signal intensity is measured as a function of repolarization time between the measurement sequences. The measured value,  $T_1 \approx 17$  s, agrees well with the results of conventional NMR measurements conducted on the same sample. b) Nutation experiment demonstrating  $rf$  intensity of 7.8 G with 100 mW of applied  $rf$  power.

## Ultrahigh Sensitivity Cantilevers

In order to achieve single nuclear spin MRFM sensitivity, we must minimize various sources of noise. The thermomechanical cantilever force noise is fundamental; its frequency independent spectral density is

$$S_{\text{th}}^{1/2} = \sqrt{\frac{4k_B T \delta\nu}{\omega_0 Q}}$$

where  $k$  is the spring constant of the cantilever,  $k_B T$  is the thermal energy,  $Q$  is the quality factor and  $\delta\nu$  is the measurement bandwidth. Clearly we can reduce  $S_{\text{th}}^{1/2}$  by fabricating a cantilever with a low spring constant. This cantilever should also be small in order to keep its resonant frequency  $\omega_0$  relatively high. We have fabricated low mass cantilevers  $60\text{ }\mu\text{m}$  long (Fig. 12a) with typical spring constant  $k \approx 0.7$  mN/m and resonant frequency  $\omega_0 \approx 2\pi \times 3000$  Hz. The UIUC, OSU and IBM-Almaden groups collaborated to fabricate a batch of such cantilevers at the Stanford Nanofabrication Facility; the actual fabrication was performed by UIUC postdoctoral researcher Trevis Crane working with the IBM group of D. Rugar. The collaboration has continued as we de-

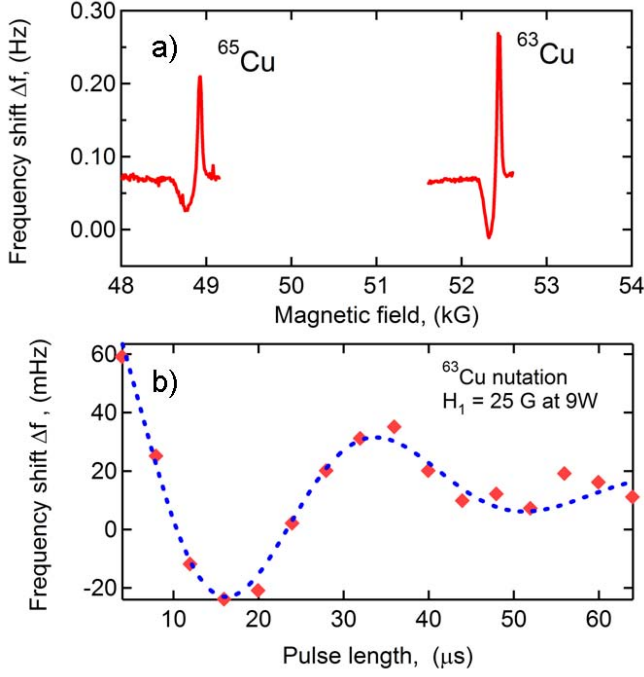


FIG. 11: a)  $^{65}\text{Cu}$ ,  $^{63}\text{Cu}$  NMRFM spectra obtained on a thin Cu foil. b) Nutation experiment on  $^{63}\text{Cu}$  spins demonstrating  $rf$  intensity of 25 G with 9 W of applied  $rf$  power.

velop the technology to use the ultrasoft cantilevers in our MRFM experiments. Using cantilevers of this type, we have demonstrated thermal force noise level as low as  $6 \text{ aN}/\sqrt{\text{Hz}}$ . These cantilevers have been used in our two electron spin sensitivity experiment.

#### Micromagnetic tips on ultrasoft cantilevers

High probe magnetic field gradient  $\nabla B$  is essential for high sensitivity MRFM. The magnitude of  $\nabla B$  defines the strength of the dipolar probe-sample interaction and should be maximized to increase the signal-to-noise ratio (SNR). This requires minimizing the dimensions of the probe magnet while maintaining high saturation magnetization and coercivity. We do this by manually mounting a SmCo microparticle on a cantilever and using Focused Ion Beam (FIB) machining to further reduce its dimensions. The characteristic dimensions of the resulting micromagnetic probe tips  $\sim 1 \mu\text{m}$  (Fig. 12c). Probe magnets such as these routinely generate field gradients exceeding  $2 \text{ G/nm}$  used, and were essential for our two electron spin sensitivity experiment. This work was done in collaboration with IBM group of D. Rugar.

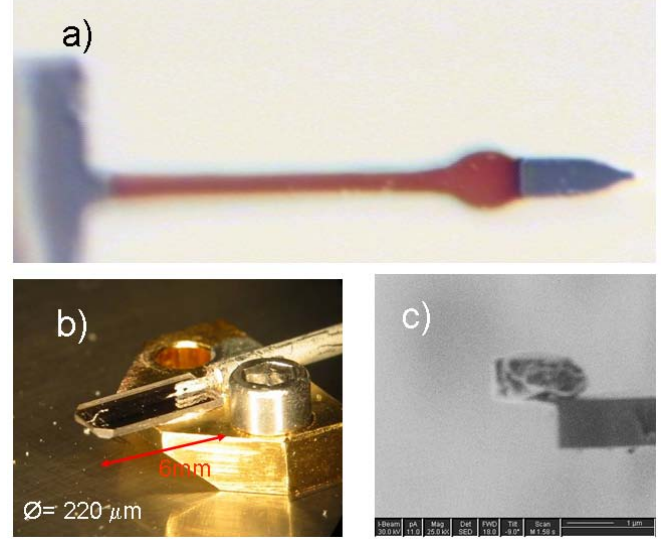


FIG. 12: Critical MRFM components. a) Ultrasoft Si cantilever developed in collaboration with IBM group of D. Rugar. Ultralow spring constant of the cantilever results in cantilever thermal noise as low as  $6 \text{ aN}/\sqrt{\text{Hz}}$ . b) Superconducting microwave resonator with a matched  $220 \mu\text{m}$  diameter coil. The coil generates magnetic field  $H_1 \sim 1.7 \text{ G}$  at applied microwave power of  $796 \mu\text{W}$ . c) FIB machined SmCo micro-magnet mounted on the end of the ultrasoft cantilever. Probe field gradients as high as  $2 \text{ G/nm}$  are routine.

#### Superconducting resonators

MRFM experiments require an intense microwave field  $H_1$  for spin manipulation. It is important to minimize the power required for a given field intensity and to confine  $H_1$  to a small volume in order to minimize the total applied microwave power, and thus the spurious heating of the experiment. One approach to this problem is to fabricate the microwave resonator out of superconducting material. We have fabricated such a resonator (Fig. 12b) based on the design developed by the IBM group of D. Rugar. The fabrication was done at OSU in collaboration with the UIUC and IBM groups. One of the challenging aspects of fabrication of such a resonator is fabrication of a  $220 \mu\text{m}$  diameter microcoil using  $25 \mu\text{m}$  Nb wire. This work was done by an OSU undergraduate student Daniel Chait for which he was awarded third prize at the OSU Denman undergraduate research forum in May 2007. Experimental testing of the Nb resonator demonstrated a resonant frequency of  $\sim 2.2 \text{ GHz}$  and a loaded  $Q$  factor exceeding 1000. We estimate that the Nb microcoil generates magnetic field  $H_1 \sim 1.7 \text{ G}$  at applied microwave power of  $796 \mu\text{W}$ . This resonator was used in our two electron spin sensitivity experiment.

## SAMPLE CHARACTERIZATION USING ESR SPECTROMETER

In this section we present results of MRFM sample characterization obtained using a Bruker Electron Spin Resonance (ESR) Spectrometer purchased with the under ARO DURIP grant W911NF-07-1-0305. These results have already been presented in the final report of the ARO DURIP grant W911NF-07-1-0305. However, while the ARO DURIP grant provided funds for instrumentation, the personnel conducting sample characterization was supported by ARO MURI grant W911NF-05-1-0414. As a result this data is also relevant to the current report.

A primary use of the spectrometer is efficient, thorough and accurate characterization of samples intended for ultra high sensitivity MRFM experiments. This section describes measurements of a particularly useful sample for this purpose. Silica contains defect sites called E' centers that carry a net electronic magnetic moment with spin  $S = \frac{1}{2}$  that are detected in an MRFM experiment. In addition to understanding relaxation dynamics of these spins, a key parameter we need to know is the spin den-

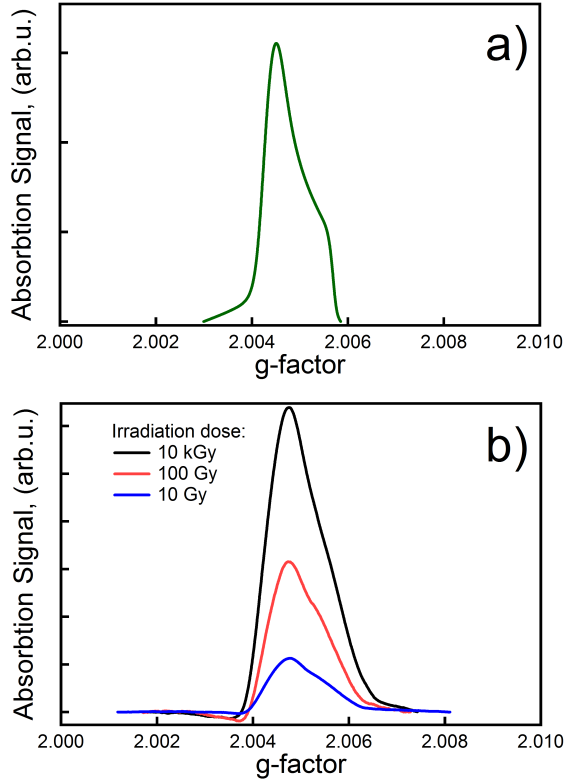


FIG. 13: Integrated ESR absorption spectra of the calibrated Wilmad sample with the known electron spin density of  $5 \times 10^{17} \text{ cm}^{-3}$  (panel a)) and the Suprasil quartz glass samples custom irradiated with 10kGy, 100 Gy, and 10 Gy doses of gamma irradiation (panel b)).

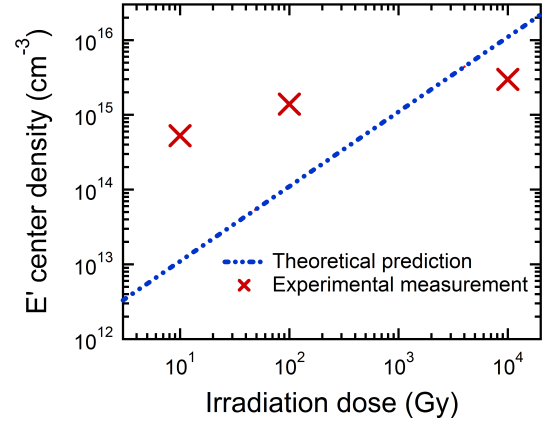


FIG. 14: E' center density in quartz as a function of the dose of gamma irradiation. The E' center density in the custom irradiated samples was measured for three doses of  $\gamma$ -irradiation (10kGy, 100 Gy, and 10 Gy) by comparing their ESR absorption with that of a calibrated Wilmad sample. The experimental data is compared with the theoretically expected numbers (dashed line). The significant discrepancy between the experimental and the theoretical data is attributed to a high density of precursor sites that existed in the samples prior to irradiation.

sity. Samples with a low spin density are of a particular interest for ultra sensitive MRFM spin detection because they allow interaction of the MRFM probe with only a few (ideally one) electron spins. Here we describe the use of the spectrometer to characterize the spin densities of a set of samples custom fabricated for MRFM experiments.

The samples were prepared by  $\gamma$ -irradiation of three Suprasil quartz crystals with different  $\gamma$ -radiation doses: 10 Gy, 100 Gy, and 10 kGy; the irradiation was performed at the Penn State Radiation Science and Engineering Center.  $\gamma$ -irradiation generates the E' defect centers we desire.

The spin density resulting from irradiation depends on the irradiation dose and the number of existing precursor sites. The latter cannot be easily measured and therefore the samples have to be experimentally characterized after irradiation. We do this by comparing ESR absorption spectra of the irradiated specimens with the spectrum of a calibrated sample with known spin density. We use a calibrated sample purchased from Wilmad-Labglass company. The 3 mm OD by 10 mm long fused glass rod was  $\gamma$ -irradiated at Cobe Labs (Lakewood, CO) resulting in a calibrated electron spin density of  $5 \times 10^{17} \text{ cm}^{-3}$ .

The integrated absorption ESR spectra of the calibrated Wilmad sample and of our custom irradiated samples was measured using the Bruker spectrometer (See Fig. 13). The total area under the measured ESR line is directly proportional to the number of electron spins in the sample. By comparing areas under the ESR lines of the custom irradiated samples with that of the calibrated

Wilma sample and using the known dimensions of the custom sample, we were able to extract the spin density resulting from the custom irradiation. The results are shown in Fig. 14 comparing the E' center density in the custom irradiated samples measured for three doses of  $\gamma$ -irradiation (10 Gy, 100 Gy, and 10 kGy) with the theoretically expected numbers (dashed line). The significant discrepancy between the experimental data and the simple theoretical expectation is attributed to the high density of the precursor sites existent in the samples prior to irradiation.

## COLLABORATIVE INTERACTIONS AND MANAGEMENT

We closely collaborate on several aspects of MRFM technology development. This approach allows faster and more efficient technology transfer thus speeding up our progress towards single nuclear spin detection. These collaborations involve both the members of the present MURI program and the members of other research institutions. The topics of the collaborative effort are listed as follows:

- **Ultrasoft cantilevers** were developed in a collaboration between OSU, UIUC and IBM. The cantilevers were fabricated at the Stanford Nanofabrication Facility by UIUC postdoctoral researcher Trevis Crane with the technical support of the IBM group of D. Rugar. The collaboration has continued while we developed the technology for use of the ultrasoft cantilevers in our MRFM experiments leading to two electron spin sensitivity.
- **Superconducting microwave resonators** were developed in a collaboration between OSU, UIUC and IBM. Superconducting microwave resonators generate high intensity microwave magnetic fields at low applied microwave power. The collaboration included development, fabrication and application of the resonators.
- **Software for statistical analysis of MRFM signal** We have adopted the software package developed by the IBM group that performs statistical analysis of the statistical polarization MRFM data that is being recorded and provides the statistical estimate of whether the acquired data is indeed a magnetic resonance signal.
- **Si nanowires for MRFM probe fabrications** We are developing Si nanowires as MRFM cantilevers. Fabrication of these nanowires presents a significant challenge. We are collaborating with Professor Lincoln Lauhon (Department of Materials Science and Engineering, Northwestern University), an expert in fabricating semiconductor

nanowires, to fabricate single crystal Si nanowires as resonant force detectors.

## Collaboration Management

Our MURI interactions and collaborations provide sharing of state-of-the-art technology for developing and enhancing ultrasensitive MRFM as described above. The interactions that enable this collaboration are achieved by frequent interaction. In addition to electronic interactions we find that face-to-face meetings are very valuable for allowing in depth discussions and interchanges:

- May 2005 OSU, UIUC and IBM team members meet at Almaden
- May 2006 OSU and UIUC team members meet at Urbana
- June 2006 OSU, UIUC, IBM, UW and Cornell teams meet at Cornell MRFM summer school
- Nov 2006 PCH visits Schwab and Cornell team at Cornell
- Dec 2006 OSU and IBM team members meet at Almaden
- June 2007 Dan Rugar meets with OSU team at OSU

## TECHNOLOGY TRANSFER AND DOD INTERACTION

One of our key goals is the fruitful application of the technical and scientific expertise that we have acquired in the course of our ultrahigh sensitivity MRFM measurements to problems of importance to DOD and the nation. In order to facilitate this technology transfer, we are collaborating with DOD, DOE and industrial labs to use ultrasensitive MRFM for Ferromagnetic Resonance (FMR) detection in submicron magnetic of interest for magnetoelectronics, magnetic field sensors and the magnetic data storage. The work is done in collaboration with E. Nazaretski and R. Movshovich from the Los Alamos National Laboratory (LANL) and J. W. Baldwin and B. Houston from the Naval Research Lab (NRL) and S. Batra (Seagate Research). This effort leverages DOE supported work (grant DE-FG02-03ER46054). The results of this technology transfer have been published in several scientific journal publications. In addition we are collaborating with J.C. Zhao at General Electric Global Research to use MRFM to microscopically image novel permanent magnet materials.

## PUBLICATIONS AND AWARDS

- "The Magnetic Resonance Force Microscope," P.C. Hammel and D.V. Pelekhov, **Book Chapter**, *Handbook of Magnetism and Advanced Magnetic*

*Materials*, Helmut Kronmüller and Stuart Parkin, eds., Volume 5: Spintronics and Magnetoelectronics, John Wiley & Sons, Ltd. ISBN: 978-0-470-02217-7, (2007).

- “High Sensitivity Magnetic Resonance Force Microscopy of Electron Spins in Silicon Dioxide,” Palash Banerjee, Yulu Che, K.C. Fong, Yuri Obukhov, Denis V. Pelekhov, P. Chris Hammel, in preparation.
- “Manipulating Spins by Cantilever Synchronized Frequency Modulation: A Variable Resolution Magnetic Resonance Force Microscope,” K.C. Fong, I.H. Lee, P. Banerjee, Yu. Obukhov, D.V. Pelekhov and P.C. Hammel, *Appl. Phys. Lett.* **93**, 012506 (2008)
- “Displacement detection of silicon nanowires by polarization-enhanced fiber-optic interferometry,” J.M. Nichol, E.R. Hemesath, L.J. Lauhon, R. Budakian, *Appl. Phys. Lett.* **93**, 193110 (2008)
- “Ion Milled Nanoscale Ferromagnetic Particle Characterization Using Cantilever Magnetometry” Ross Steward, 2nd prize at Richard J. and Martha D. Denman Undergraduate Research Forum, Ohio State University, May 17, 2006
- “Fabrication and Characterization of Superconducting Microwave Resonators for Subatomic-Scale Spin Resolution,” Daniel Chait, 3rd prize at Richard J. and Martha D. Denman Undergraduate Research Forum, Ohio State University, May 16, 2007.

#### Awards

- Prof. P.C. Hammel named **Fellow of the AAAS** 2005.

#### Invited Talks

- “Ultrasensitive Electron Spin Resonance with the Magnetic Resonance Force Microscope,” presented at *EPR 2005*, Columbus, OH, on 7 September 2005.
- “The Magnetic Resonance Force Microscope: A New Tool for High Resolution Materials Studies,” presented at the 135th Annual Meeting of *The Minerals, Metals & Materials Society (TMS)*, San Antonio, TX, March 13, 2006.
- “The Magnetic Resonance Force Microscope: A New Tool for High Resolution Materials Studies”

presented at the *Summer School on Magnetic Resonance Force Microscopy*, Ithaca, NY, June 23, 2006.

- “Ultrasensitive Magnetic Resonance Detection with Micromechanical Cantilevers” presented at the *Symposium on Nonlinear Dynamics of Nanosystems* at Chemnitz, Germany, August 29, 2007.
- “Force-Detected Scanned Probe Magnetic Resonance Microscopy,” Colloquium presented at Miami University of Ohio, October 19, 2005.
- “Ultrasensitive Magnetic Resonance Detection with Micromechanical Cantilevers” Seminar presented at the Institute for Solid State Research of the Leibniz Institute for Solid State and Materials Research, Dresden, Germany, September 1, 2006.
- “Submicron Magnetic Resonance Imaging Using Scanned Probe MRFM,” Solid state seminar presented at the Department of Physics, Cornell University, Ithaca, NY, 14 November 2006.
- “Submicron Magnetic Resonance Imaging Using Scanned Probe MRFM,” presented at the Los Alamos National Lab Materials Colloquium, Los Alamos, NM, 21 February, 2007
- “Scanned Probe Magnetic Resonance Imaging for Magnetoelectronics,” Colloquium, Department of Physics, Kent State University, Kent, OH, 12 April, 2007

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